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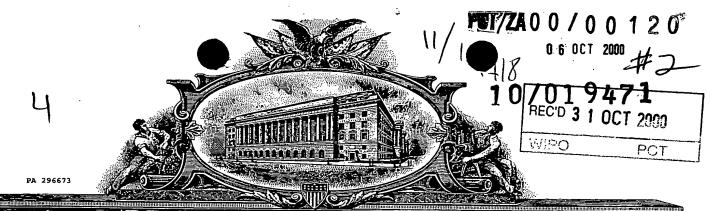
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APPLICATION NUMBER: 60/142,382

FILING DATE: *July 06, 1999* 

# PRIORITY DOCUMENT

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B a request for filing a PROVISIONAL APPLICATION under 37 CFR 1.53(c).

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INVENTOR(S)/APPLICANT(S)											
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TITLE OF THE INVENTION (280 characters max)											
HIGH TEMPERATURE METATHESIS PROCESS											
CORRESPONDENCE ADDRESS											
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STATE	VA		ZIP CODE	22	202			COUNTRY	USA		
ENCLOSED APPLICATIONS PARTS (check all that apply)											
х	Specifica- tion	No. of Pages		8			Small I	Entity Statement			
х	Drawing(s)	No. of Sheets		1		Other	ther (specify):				
METHOD OF PAYMENT (check one)											
Х	A check in the amount of \$150 to cover the filing fee is enclosed.					•	Check No.  14491 PROVISIONAL FILING FEE				
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[X] Additional inventors are being named on separately numbered sheets attached hereto.

The invention was made by an agency of the United States Government or under a contract with an agency of the United States Government.

[X]

Yes, the name of the U.S. Government agency and the Government contract number are:

Respectfully submitted,

Benoit Castel

July 6, 1999

Reg. No. 35,041

PROVISIONAL APPLICATION FILING ONLY

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Rev 2 - 5 July 1999

#### HIGH TEMPERATURE METATHESIS PROCESS

This invention relates to a high temperature metathesis process. In particular, the invention relates to the optimisation of the high temperature metathesis process to improve selectivity for a desired product range.

The applicant is aware that olefins in the  $C_8$  to  $C_{14}$  may be used as detergent and plasticizer precursors as well as for alkylation of benzene, and that  $C_{15}$  to  $C_{18}$  olefin ranges may be used as drilling fluids and drilling fluid precursors, amongst other uses.

Conventional thinking was that linear olefins may be used to produce linear alkyl benzene and linear oxo-alcohols which could be used to produce detergents and plasticizers which were believed to be both bio-degradable and suitable for their intended purpose. Thus, previously efforts were concentrated on producing linear oxo-alcohols and lineal alkyl benzene, and thus efforts were focused on linear olefins from which these could be made.

Recently, however, a new wave of thinking has lead to the belief that non-linear oxo-alcohols as well as non-linear alkyl chain alkyl benzene could be used alone or together with their linear counterparts for the production of said detergents and plasticizers. In particular short chain branched olefins are believed best sulted to produce such non-linear products. Thus, recent efforts have concentrated on the delinearization of the linear olefins in order to use such olefins in the production of the non-linear products.

Rev 2 - 5 July 1999

Surprisingly, after extensive research, the applicant has found that a peculiar olefin composition in the  $C_9$  to  $C_{18}$  range, having both linear and non-linear olefins may be made by metathesis of Fischer-Tropsch olefins in the  $C_5$  to  $C_{15}$  range.

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Thus, according to a first aspect of the invention, there is provided a high temperature metathesis process for the metathesis of Fischer-Tropsch olefins in the C<sub>5</sub> to C<sub>15</sub> range, said metathesis process including the step of subjecting a Fischer-Tropsch olefin feedstock in the C<sub>5</sub> to C<sub>15</sub> range to metathesis reaction conditions, said olefin feedstock including mono-methyl branched olefins.

The high temperature metathesis process may be carried out at a temperature of between 300°C to 600°C.

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Typically the high temperature metathesis process is carried out at a temperature of between 450°C and 550°C.

The operating pressure of the high temperature metathesis process may be between 1 and 30 bar, or even higher.

The high temperature metathesis process may use a tungsten or molybdenum based catalyst, for example, WO3 or MoO3, supported or unsupported, with or without co-catalysts. The support can typically be SiO2, Al2O3, ZrO2, TiO2, or mixtures thereof.

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Rev 2 - 5 July 1999

The high temperature metathesis process Fischer-Tropsch olefinic feedstock in the C<sub>5</sub> to C<sub>15</sub> range may include linear alpha olefins, monomethyl branched olefins, paraffins, dienes, aromatics, and the like.

Typically, the Fischer-Tropsch olefinic feedstock includes one or more olefins selected from the  $C_5$  to  $C_8$  range.

The product of the high temperature metathesis process may include one or more mono-methyl branched olefins in the C<sub>9</sub> to C<sub>10</sub> range.

The product of the high temperature metathesis process may include one or more linear olefins in the  $C_{\rm B}$  to  $C_{\rm 18}$  range.

The product of the high temperature metathesis process may include one or more mono-methyl branched olefins and one or more linear olefins in the  $C_9$  to  $C_{18}$  range. The olefins of the product may be internal olefins.

The product of the high temperature metathesis process may be used in the production of alkyl benzene, plasticizers, detergents, drilling fluids, and the like, having both a linear fraction and a branched fraction (for alkyl benzene the alkyl chain is branched or linear).

Typically, the branched fraction will be mono-methyl branched.

However, the branching may be di-methyl, ethyl, and/or propyl.

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Rev 2 5 July 1999

According to a second aspect of the invention, there is provided a high temperature metathesis process for the metathesis of olefins in the C<sub>5</sub> to C<sub>15</sub> range, said metathesis process including the step of subjecting an olefinic feedstock in the C<sub>5</sub> to C<sub>15</sub> range to metathesis reaction conditions, the process including the recycling of a part of the product of the metathesis reaction to the reaction to increase the selectivity for a desired product range.

The olefinic feedstock may be a Fischer-Tropsch olefinic feedstock

in including mono-methyl branched olefins.

Typically, the olefinic feedstock includes one or more olefins in the  $C_{\text{\tiny B}}$  to  $C_{\text{\tiny B}}$  range.

Where the desired product range includes olefins in the  $C_9$  to  $C_{18}$  range, the process includes a separation stage wherein a recycle fraction in the  $C_5$  to  $C_8$  range is separated from the product and recycled to the reaction.

The quantity of recycle in the feedstock may be selected to provide a .C9 and higher selectivity of above 50%.

Generally, the quantity of recycle in the feedstock is selected to provide a C<sub>9</sub> and higher selectivity of above 50%.

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Rev 2 - 6 July 1999

Typically, the recycle makes up between 20% and 80% of the reaction feedstock.

Usually, the recycle makes up between about a third and three guarters of the reaction feedstock.

The total yield of high temperature metathesis process product in the  $C_9$  to  $C_{18}$  range is above 40%.

Typically, the total yield of high temperature metathesis process product in the C<sub>9</sub> to C<sub>18</sub> range is about 50%.

The total feedstock conversion of the high temperature metathesis process of the invention is typically in the range of 60% to 90%, usually about 80%.

The ratio of linear to branched high temperature metathesis process products is typically greater than 1:1.

Usually, the ratio of linear to branched high temperature metathesis process products is greater than 2:1.

Generally, the ratio of linear to branched high temperature metathesis process products is about 3:1.

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Rev 2 - 5 July 1999

The branching of the high temperature metathesis process products is predominantly mono-methyl branching, although some di-methyl, ethyl, and/or propyl branching may also be present.

The product of the high temperature metathesis process may be used in the production of alkyl benzene, plasticizers, detergents, drilling fluids, and the like, having both a linear fraction and a branched fraction (for alkyl benzene the alkyl chain is branched or linear), the ratio of linear to branched fractions being related to the ratio of linear to branched high temperature metathesis process products used in their production.

The invention will now be described, by way of non-limiting illustration only, with reference to the accompanying line diagram.

In the diagram, reference numeral 10 generally indicates a high temperature metathesis process broadly in accordance with the invention.

The process 10 includes a reactor 12 operated at between 450°C and 550°C and at an operating pressure of between 1 and 30 bar. A Fischer-Tropsch-olefinic-feedstock 14 including mono-methyl branched olefins, is fed into the reactor 12. The feedstock 14 includes olefins in the C<sub>5</sub> to C<sub>9</sub> range.

Usually the feedstock 14 will be purified of oxygenates which may poison the catalyst by extractive distillation (not shown), prior to being fed to the reactor 12.

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Rev 2 - 5 July 1999

The reaction product 16 includes both linear and branched internal olefins in the  $C_2$  to  $C_{18}$  range.

The reaction product 16 is fed to a separator 18 where it is cut into a light product stream 20 including  $C_2$  to  $C_4$ , a recycle stream 22 including  $C_5$  to  $C_6$ , and a heavy product 24 including product in the desired  $C_8$  to  $C_{18}$  range.

The recycle stream 22 is combined with the feedstock 14 to form the total feedstock of the reactor 12

The recycle stream 22 is between a third and and three quarters of the feedstock 14.

The total yield of heavy product stream 24 is about 50%, while the feedstream 14 conversion is about 80%, with a selectivity for C<sub>0</sub> to C<sub>18</sub> of about 60%.

The ratio of linear to branched product in heavy product stream 24 is about 3:1.

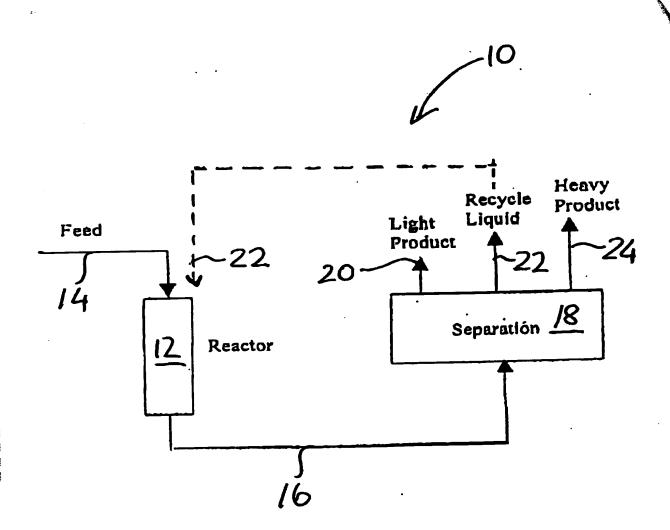
Rev 2 - 5 July 1999

The applicant believes that it is an advantage of the invention as illustrated, that the high operating temperatures result in a high degree of resistance to poisoning of the metathesis catalyst by feedstock components, such as branched olefins, dienes, aromatics, and the like.

The applicant believes that it is a further advantage of the invention as illustrated that by recycling a cut of the product which is below the desirable carbon length range, high selectivity to desired products is achieved..

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